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Introduction

The Heavy Element Facility[1] is a cold war legacy facility at Livermore National Laboratory. The facility's mission has varied over its lifetime, but operations included the preparation of radioactive heavy element tracers used in underground nuclear weapons testing and the conduct of a heavy element research program. It is a one story concrete masonry structure constructed in several phases between 1955 and 1981.

In 1993, a seismic re-evaluation of the facility determined that portions of the building did not meet the PC-2 requirements applicable to it. A seismic upgrade evaluation determined it was not practical to upgrade the facility to support continued programmatic operations. It is now maintained in a storage mode awaiting Department of Energy disposition. In this mode the operations are limited to (1) storage of radioactive material from previous operations, (2) clean-up and decontamination of facility work areas and equipment, (3) removal of contaminated systems and enclosures, (4) facility maintenance, (5) removal of radioactive materials from the facility, (6) characterization of the waste generated by these activities, (7) surveillance activities and (8) security.

An important part of the facility's storage function is provided by underground storage vaults. These are embedded in a massive reinforced concrete block whose top is at the building interior's floor level. The inventory in these vaults is limited to solid forms of transuranic isotopes and other radioactive isotopes stored with double or triple containment. The vaults may be accessed infrequently for surveillance or on occasion for removal of inventory to other facilities.

As part of maintaining this storage function until final disposition, the safety of the underground storage system was reevaluated using guidance in DOE standard[2] DOE-STD-1027-92. An overview is presented here to highlight important considerations in the evaluation of an older safety system. Special effort is directed to effects of aging when screening for failure modes, energy sources and initiating events. Processes influencing aging include radiation, transmutation of radioactive elements within the solid material forms, and generation of helium gas from alpha decay. Affected objects include the radioactive material solids and the containers, including the o-ring of the outer container.

Seismic events were identified as the dominant concern after a screening of potential damage initiators. The vault embedment and cover plate protect the container barriers from major damage during an earthquake. But combined with aging effects, the earthquake may cause some narrow cracks in the barriers. Leakage paths proceed outward through the concentric containers, the vault, and the HEPA filter exhaust system, which itself may be damaged by the earthquake. The prior buildup of He pressure in the containers, as the result of radioactive decay, may provide energy and a motive force to drive fine particles through the leakage paths. The release by this physical process is extremely limited. The analysis takes no account of attenuation by fallout or plateout. The potential off-site consequences remain far below the emergency planning limit for the site.

The potential damage and release that can result when an underground storage rack is raised for surveillance or removal of a container is also evaluated. In this case the damage caused by a concurrent earthquake may be more severe and the release path to the environment is reduced. He gas still plays a large part in the release mechanism. The off-site consequences are substantially higher than from an earthquake during storage mode even though the inventory involved is smaller, but they are still below the emergency planning limit for the site, of 5 rem committed effective dose equivalent (CEDE) at the site boundary.

Storage Description

The materials are stored in cylindrical metal containers placed in storage vaults in a massive below-grade embedment. The embedment is essentially a monolithic reinforced concrete block larger than 12 x 12 x 12 feet, with recessed pipes set in the concrete block as storage vaults (Fig. 1). A sump provides collection of any water such as fire-suppression sprinkler water. The pipe vaults are covered by massive cover plugs, and the recessed pipe array is covered by steel plates at the building grade level. The vaults are not leak-tight but a connection to the building HEPA exhaust provides a control of airflow direction during normal storage. Other features for physical security and safeguards do not interfere with the storage system features just described, and are not included in the safety analysis.

The radioactive materials are typically in multiple concentric cylindrical containers within the vault system. A typical radioactive unit is a solid oxide or metal piece, in a small metal container inside a juice can inside the storage container in the vault. The innermost container types include screw-top cans and soldered cans. The juice cans are typically crimped as a closure, and the storage containers have a bolted closure with a single o-ring seal. The series of cans, the pipe vault, and the vestibule of the pipe array provide a series of volumes with at most a very limited connection from one to the next.

Initiating Event Screening

A broad survey of possible failure modes, out-of-specification conditions, energy sources, and initiating events was conducted to find possible release scenarios during the storage mode.[3]

Initiating events surveyed include accidents internal and external to the building, flooding, fire, and earthquake. Additional failure modes were identified arising from the unique nature of the stored materials. When initially sealed, these materials consisted of isotopically pure solids or

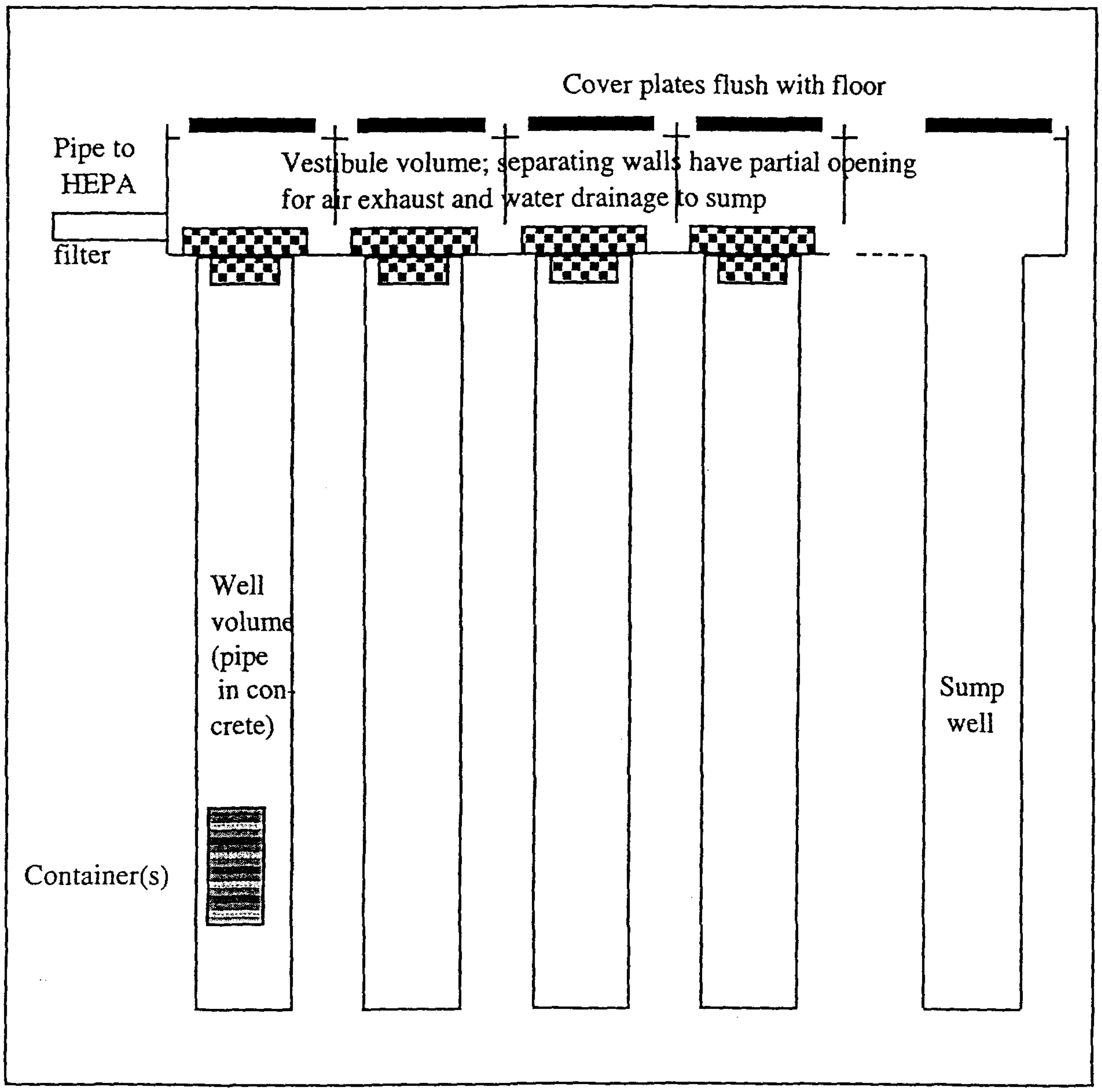

Fig. 1. Schematic of the underground storage vault system.

solids of known isotopic content. Representative stored pure isotopes are ^{232}U (72 years half-life) and ^{244}Cm (18.1 years half-life) in quantities of up to one or two grams of the radioactive element. These isotopes are not fissile isotopes, but they have relatively high specific radioactivity because of the short half-lives. During the time (10-40 yrs.) to the present, typically about one-quarter to three-quarters of the radioactive atoms in a sample have decayed via alpha decay to another element. This may lead to substantial change to the solid structure of the radioactive piece, possibly generating powders, and it leads to the generation of a substantial amount of helium gas. The extent of leakage or solid diffusion of the He gas is not known, hence a range of logical possibilities is considered. The helium gas may leak out or it may build up pressure inside the innermost container, ready to cause a leak when the container is mechanically damaged. Conceivably if a leak occurs suddenly when a threshold of internal pressure is reached, then some radioactive powder could be conveyed out of the concentric containers series into the below-grade vault area where it may settle. The alternate potential conditions of no leak or prior leak before an earthquake are considered in the response analysis below.

The o-ring in the outer container may deteriorate due to aging, a radiation field, or slightly elevated temperatures. This could cause a very narrow flow path for gases from the interior.

Some failure modes are screened from further consideration on the basis of their absence or limited effects. Process gas lines to the storage rooms have been shut off. A natural gas line for building heating enters from the opposite side and does not pass near the storage room. External vehicle traffic near the building is minimal. A flooding event would leave extensive time for recovery before consequences such as rust develop. The potential for a fire is limited by a fire safety program, minimal combustibles loading, a sprinkler system, and the administrative exclusion of hydrocarbon-fueled trucks and forklifts from the building. The location of the stored containers within a concrete monolith and covered by a massive concrete and steel cover provides substantial thermal isolation from a fire in the building. A severe earthquake provides the greatest potential challenge to the storage system, and was analyzed in more detail in its effects and consequences.

As noted above, a 1993 seismic re-evaluation of the facility determined that portions of the building did not meet the PC-2 requirements applicable to it. Thus for the below-grade storage system, a collapse of some portions of the building is assumed as possible. A structural analysis of the storage system (by Steven Lu, available as Appendix B of Ref. 3) showed that the storage vaults will not be damaged by direct effects of the earthquake and the cover plates will not be broken or penetrated by falling objects from the structures or equipment in the building. In the earthquake consequence analysis for the vaults, it is assumed that the building HEPA exhaust system may be stopped or its connecting ducts may be breached.

In summary the energy sources of fire and falling objects cannot reach the stored containers. The energy of direct earthquake shaking cannot damage the containers substantially. Thus fire or earthquake alone will not cause a release consequence. That leaves the energy of stored compressed helium gas, either acting alone to cause an initial crack or acting together with an earthquake event.

A list of possible failure modes and events after screening is shown in Table 1. The fire and water flood events as single events are screened out but are considered as a later event after a prior helium gas-initiated leak from the container to the vault, and when caused by an earthquake.

The leading hazards, from aging and earthquake, are considered further in this paper. Other hazards are included in Ref. 3.

Table 1. Possible associations of events with scenario elements.

Scenario elements --> / Components/Events	Create powder	Suspend powder	Leak path	Motive force for leak
Stored materials	✓			
Aging - solid deterioration	✓			
Aging - helium generation		✓	✓	✓
Aging of O-ring			✓	
Fire			✓	✓
Water flood		✓		✓
Earthquake (may include a fire or flooding)		✓	✓	

Consequence Analysis

Seismic event during in-vault storage

Helium gas is a potential source of pressure in a small interior container and of motive energy for transport of any fine particulates. The pressure and the motion effects depend on the successive container gas volumes. This section discusses the creation of helium gas, the development of a powder component in the stored solids, the cracking of the innermost small container, and the mixing and entrainment of powder through the succession of volumes. (Alternately the closure could leak helium slowly or allow diffusion through the solid, thus avoiding cracking and entrainment. This alternative is not evaluated further.) The innermost container's crack development could occur before an earthquake, allowing some powder to be entrained and to be settled out in the pipes and storage vestibule, available for resuspension in an earthquake. Subsequently generated helium would move through the cracks without entraining any additional powders. Alternately, the innermost container could be pressurized near the failure threshold, and be pushed over the failure threshold by the jostling from the earthquake. This latter event is unlikely but will be evaluated. The ducts leading to the HEPA filter are conservatively assumed to be ruptured by the earthquake, thus shortening the path to the external environment.

The amount of helium gas is directly proportional to the amount of alpha decays. We consider examples of ^{244}Cm and ^{232}U, starting at 2 grams each. After a storage time interval of one half-life ($t_{1/2}$ = 18.1 and 72 years respectively), one gram has decayed. For ^{244}Cm, this one gram is approximately 1/244 mole, producing 1/244 = 0.0041 mole of helium and a like molar quantity of ^{240}Pu ($t_{1/2}$ = 24,000 yrs.). For ^{232}U, the one gram is 1/232 mole. Further decays (all rapid compared to the storage time) in the ^{232}U decay chain result in a total of six helium atoms for each ^{232}U atom decayed, thus 6/232 = 0.026 mole of helium. The latter would occupy 580 cm^3 at 25°C and 760 mm Hg pressure (STP).

Some of the small interior containers have a gas volume on the order of 2 cm^3. With ^{232}U, adding in 0.026 mole of helium in the inner small capsule would raise the pressure to 290.5 atmospheres or 4270 psi. If the seal area on the perimeter of the lid is 5% of the total lid area, the stress on the seal would be 20x4270 = 85 ksi on average, with possible stress-riser geometrical details locally. For the case of 1 g ^{244}Cm decayed from the 2 g initial ^{244}Cm, the stress at the seal would be 13.8 ksi. The yield stress of some solder materials is well below 10 ksi (see section "Low-melting Metals and Alloys" in Sec. 6 of Ref. 4), hence at some point in the helium buildup the lid closure might well yield and crack. The amount of helium at that stage would be less than the 0.026 mole cited above. For soldered closures, a yield pressure of 4 to 8 ksi for the closure means failure at a helium production level of 0.0015 to 0.0030 moles.

The stored solid by that time may well have some powder component, due to the radionuclide decays which involve solid lattice damage and elemental transmutation leading to changed chemical valence properties. If a sudden crack and pressure release occur, then some powder would be entrained. A fraction of 0.001 of the radioactive inventory is used for the amount entrained out of the first container. This is based on the generic fraction used in DOE STD 1027-92 Appendix A. It is conservative compared to values for the aerodynamic entrainment of powders or for the airborne release from fires on contaminated materials as discussed[5] in DOE HDBK 3010-94, Sections 4.4.4.1 and 4.4.1.1 respectively.

After a puff release from the innermost capsule, there is a succession of containers one within another (see Fig. 2). We assume they are all susceptible to leaks, whether through a crimped seal on a can or through an aged o-ring seal. Each successive container has a net air volume V_i, which converts to an air mass m_i in moles. For an initial added mass of m_{he} moles of helium, eventually m_{he} moles of gas will exit the breaches to restore one atmosphere pressure. We assume mixing of the released fine particulates in each volume V_i, for $i \geq 1$. The fraction of particulates exiting a container with the gas will be $m_{he}/(m_i + m_{he})$ of the amount entering, or $V_{he}/(V_i + V_{he})$ where the gas volumes are those at STP.

The radioactive material items such as small capsules are in most cases stored in a juice can within a storage container, but some items are stored in the storage container. We neglect additional intermediate packaging such as polyethylene bags for the analysis. The case of an item directly in the storage container has a higher release fraction than the case with an intermediate juice can. The quantity of helium at the time of a crack breach plays a major part in the quantity released. For the storage mode we consider the full 0.026 mole of helium developed by an initial 2 g of ^{232}U at one half-life. This is quite conservative compared to the likely helium levels at the time of a breach. As a case more modestly above the likely average helium level at breach, we consider the ^{244}Cm example described above, with 0.0041 mole of helium.

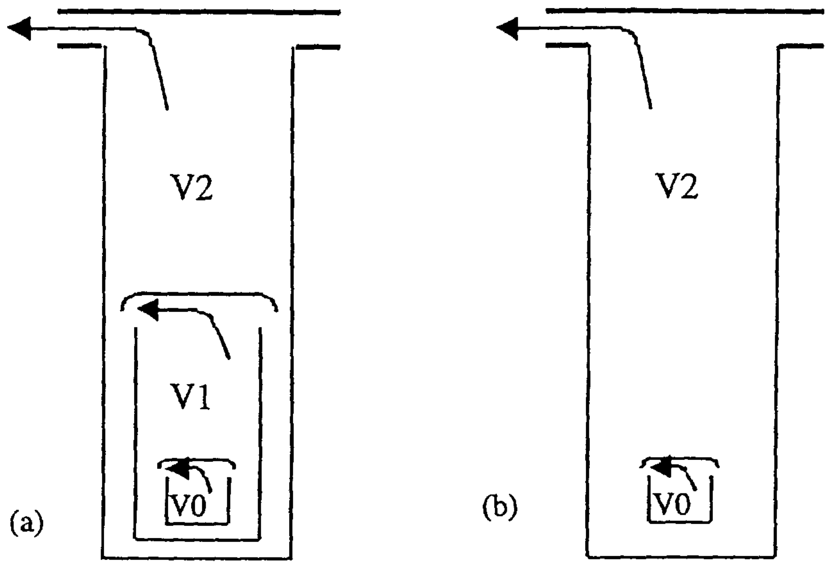

Figure 2. Schematic of container release scenario during in-vault storage. The release is initiated by a crack opening in the innermost container. Release is shown as far as the storage pipe vault. (a) Storage within an intermediate juice can. (b) Storage without the an intermediate juice can.

First we examine the case where a small capsule is stored directly in a storage container, without a juice can as well. The storage container volume is on the order of $V_2 = 3000$ cm^3. For the ^{232}U example, the fraction of incoming particulates exiting is 580/(3000 + 580) = 0.162. Thus 1x10^{-3} x 0.162 = 1.62x10^{-4} fraction of the radioactive material is moved as far as the vault volume. Next the vault volume is about 12,000 cm^3. Then the factor for further exiting is 580/(12000 + 580) = 0.046. Thus the fraction of the radioactive material moved to the vestibule is 1.62x10^{-4} x0.046 = 7.45x10^{-6}. A fraction 1.55x10^{-4} remains in the vault and settles out to be available for later accident scenarios. Of the amount reaching the vestibule, an undetermined fraction of it settles out and is available for later scenarios, and the other fraction is carried off to the HEPA filter system.

For most materials, a juice can or similar can is used as an intermediate storage container. This has a volume of about $V_1 = 1500$ cm^3. Then the net air volume of the outer container is reduced to $V_2 = 1500$ cm^3. Then with this additional can is the sequence, for the ^{232}U example the fraction released to the vault or to the vestibule is reduced by a factor 0.48 from 7.45x10^{-6} to 3.58x10^{-6}.

For the ^{244}Cm case with its smaller helium volume of 0.0041 mole or 92 cm^3 at STP, first for a case with a small capsule stored in a storage container, the fraction of the stored material reaching the vault is 3.0x10^{-5}, and the fraction reaching the vestibule is 2.3x10^{-7}. For a ^{244}Cm case with a juice can as an intermediate storage container, the fractions reaching the well or the vestibule are reduced by a factor of 0.112 to 3.3x10^{-6} and 2.6x10^{-8} respectively.

Now if an earthquake occurs and the initial container has not cracked yet, then such a crack could occur, pushed over the failure threshold by the jostling from the earthquake. Then the release is as calculated above for a pressure-induced crack. The HEPA system may be stopped or its ducts damaged by the earthquake, so we assume that the fraction reaching the vestibule can be released to the outside air. Alternately if the container has cracked prior to the earthquake, then that release does not occur concurrently with the earthquake. But a resuspension of particulates deposited in the vaults-array vestibule could take place. The resuspension is a small fraction, so this alternative has a much smaller release than the other alternative of initial release.

The specific activity of ^{232}U is 21.4 Ci/g, and of ^{244}Cm is 80.9 Ci/g. Thus an initial 2-gram amount of U-232, when decayed to 1 gram, is 21.4 Ci and has daughter products at 21.4 Ci each. A fraction 7.45×10^{-6} reaching the vaults' vestibule would be 1.59×10^{-4} Ci of ^{232}U and each daughter. The consequences of a release of this size may be evaluated using a simple Gaussian model such as HOTSPOT[6] and results in a 50-yr. CEDE at the closest Laboratory boundary of 5.2×10^{-2} rem for the ^{232}U example. The facility-specific results, taking account of the ages and quantities of the stored materials now and in the future storage periods, are lower than in the 2-gram examples. The packaging details (e.g., plastic bags, deposition on packaging materials surfaces), ignored in this model, would further reduce the estimates.

Seismic event during inventory removal

The time during which any vault is open and the rack, upon which the storage containers are located is raised, is kept to a minimum. However, there is still the possibility of an earthquake occurring while the vault contents are exposed. Each rack is approximately 12 ft. long and has shelves spaced about 1 foot apart. There is, at most, a single final storage container per shelf. Thus removal of a container from the lowest shelf requires lifting the rack until the top shelf is as much as 11 ft above the floor. If an earthquake should occur at this time, or even after the rack had been lowered but a container is still in the area containing the vaults, damage to a container is credible.

The model used to describe this release event is similar to that for the in-storage event. However, the driver for the movement of powder from the containment to the bay must consider the possibility that something, e.g., concrete spalling from the walls, parts of the bridge crane mechanism, part of the roof structure, will impact the container, partially crushing it. The reduction in volume of the containers compresses the air, or other gas, sealed in the container when it was put into storage. The overpressure in the container thus is the sum of the helium pressure generated by decay and the air compression.

The details of the calculation depend on the specific packaging being considered and the sequence of events being assumed. For instance, assume that the primary and secondary container seals have previously leaked rapidly enough to have delivered 0.1% of the radioactive material to the secondary containers, then leaked to the tertiary container, where it has settled out. See Figure 3. An earthquake resuspends 1% of the material in each container. Falling debris from the building structure then crushes the secondary and tertiary containers to a fraction $(1-\beta)$ of their original volumes. The resuspended radioactive material in the containers is expelled into the room by the overpressure caused by helium and by the reduction in containment volume of the secondary and tertiary containers.

The prior leakage of material from the primary container is driven by the helium overpressure only. During this leakage, 0.1% of the inventory (I) is delivered to the secondary container by an amount of helium V_{he} generated to that time. The airborne material and helium are assumed to mix uniformly with the air in the secondary container. This mixture then leaks into the tertiary container until the overpressure due to the helium is the same in the two volumes (secondary container V_1 and tertiary container V_2). This moves a fraction $[V_2/(V_1 + V_2)] \cdot [V_{he}/(V_1 + V_{he})]$ of the 0.1% airborne inventory into the tertiary container. All of the entrained radioactive material settles on available surfaces in the secondary and tertiary containers. We assume that the released and settled material has the same composition as the original material, so its radioactive content over time remains 0.1% of the total inventory.

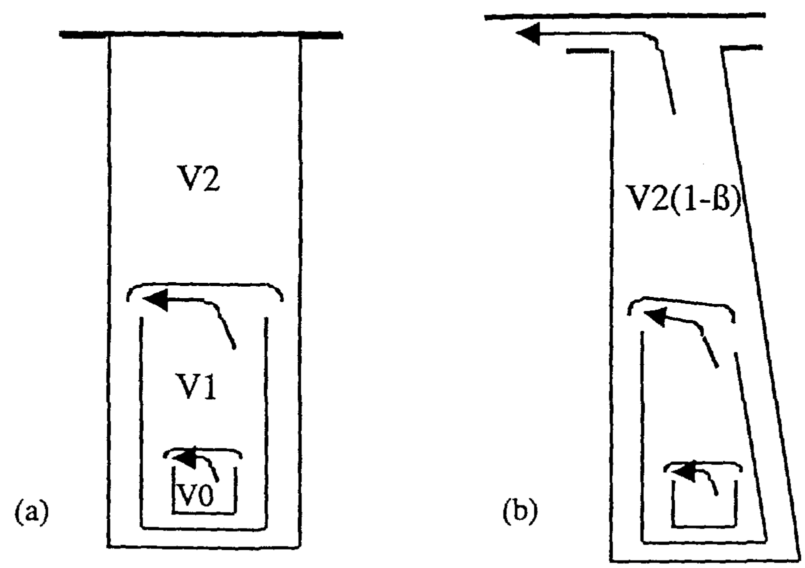

Figure 3. Schematic of release scenario during inventory removal if an earthquake occurs. (a) Prior to the removal, the innermost container is assumed to undergo a release event, while the tertiary storage container does not leak. (b) During the removal, if an earthquake occurs, the previously transported material undergoes some resuspension. The container is assumed to be damaged, and release is driven by the expelled gas volume and accumulated helium.

The storage vault is then opened and the rack withdrawn, exposing the storage containers to possible impact. A severe earthquake occurs and a container is disturbed, first by shaking, then by the impact of some external debris that partially crushes the container. The shaking is estimated to cause the resuspension of 1% of the settled material in both the secondary and the tertiary containers. A volume V_{he2} of helium generated up to that time is distributed in the secondary and tertiary containers in proportion to the container volumes.

Considering first the secondary container, the volume of gas at STP remaining in the container is $[V_1/(V_1 + V_2)] \cdot V_{he2} + V_1$. Assuming that both secondary and tertiary containers are crushed, losing a fraction β of their original volumes, the total volume of gas exiting the assembly from the secondary container is $\beta \cdot V_1 + [V_1/(V_1 + V_2)] \cdot V_{he2}$ and the fraction of the entrained radioactive material carried with it is the ratio of the exiting volume and the volume contained in the container prior to the seismic event, expressed as:

$[\beta + V_{he2}/(V_1 + V_2)]/[1 + V_{he2}/(V_1 + V_2)]$.

The radioactive material delivered to the tertiary container from the secondary container upon crushing is therefore given by 0.1% of the original inventory (I), modified by the resuspension factor 1%, and then by the volume ratio above, or:

$$I_1 = 1 \times 10^{-5} \, I \times [\beta + V_{he2}/(V_1 + V_2)]/[1 + V_{he2}/(V_1 + V_2)]$$

This assumes that nearly all of the material carried to the secondary container remains in the secondary container, i.e., it does not subtract the material settled in the tertiary container from the secondary container contents.

At the same time, the material leaked into the tertiary container before the seismic event and settled out is resuspended. This quantity is

$$I_2 = 1 \times 10^{-5} \, I \bullet [V_2/(V_1 + V_2)] \times [V_{he}/(V_1 + V_{he})].$$

Additional material from the primary container may be neglected at this stage because the driving gas amount there is very small, $V_{he2} \bullet V_0 / (V_0 + V_1 + V_2)$. Allowing all of the radioactive material exiting from the secondary container to mix in the tertiary container before release to the environment is a conservative assumption. The total inventory suspended in the tertiary container is then $I_1 + I_2$.

This inventory is suspended in a total volume of gas that is approximated by:

$$V_t = V_{he2} + \beta V_1 + V_2.$$

As the tertiary container is crushed, losing a fraction β of its volume, a volume of gas that is equal to the volume of the helium plus the fraction β of the two container volumes is released, and the ratio of this volume to V_t is the fraction of the entrained inventory released from the tertiary container. The ratio is

$$f = [V_{he} + \beta(V_1 + V_2)]/[\, V_{he} + \beta V_1 + V_2]$$

The overall release to the environment is therefore:

$$\begin{aligned} R &= f \bullet (I_1 + I_2) \\ &= 1 \times 10^{-5} \, I \bullet \{[V_{he2} + \beta(V_1 + V_2)]/[\, V_{he2} + \beta V_1 + V_2]\} \bullet \\ &\qquad \{V_2 \bullet V_{he}/[(V_1 + V_2) \bullet (V_1 + V_{he})] + [\beta(V_1 + V_2) + V_{he2}]/\,[V_1 + V_2 + V_{he2}]\} \end{aligned}$$

For a total inventory at the time of the earthquake of I, if the helium volume at that time V_{he2} is 0.05 V_1, the helium volume V_{he} is approximated by V_{he2}, the crush factor β is 0.5, and the volumes V_1 and V_2 are equal, then the total release to the environment in this scenario is 3.6×10^{-6} I. This low release fraction is comparable to the low release fraction found for the in-storage scenario, and results in a low off-site dose. When β is as large as 0.5, the release result is relatively insensitive to V_{he2} and is sensitive to β. There is a substantial margin between the off-site doses found and the site's emergency-response criteria, thus the margin allows for considerable variation in the storage system's response parameters.

Summary

The safety of the underground storage vault system of the LLNL Heavy Element Facility was reevaluated based on current criteria. The system design provides massive protection and a redundant series of containers. Some effects of aging were identified. The consequences of aging and an earthquake event are found to be small compared to the site's emergency-response criteria. This is a result of the massive nature of the storage vault system, which protects from direct effects, and of the design with multiple concentric containers, which mitigates the effects of aging-driven release events.

References

1. "Safety Analysis Report for the Heavy Element Facility (Building 251)," Lawrence Livermore National Laboratory, UCRL-AR-113377 Rev. 1DR, submitted to USDOE, May 2, 2001 (limited distribution).

2. U.S. Department of Energy Standard 1027-92, "Hazard Characterization and Accident Analysis Techniques for Compliance with DOE Order 5480.23, Nuclear Safety Analysis Reports," U.S. Department of Energy, Washington, DC, 1992.

3. W.J. O'Connell, "Hazard Analysis for B251 Underground Storage Vaults," Lawrence Livermore National Laboratory, UCRL-ID-140567, September 30, 2000 (limited distribution).

4. T. Baumeister and L.S. Marks, "Standard Handbook for Mechanical Engineers," 7th Ed., p. 6-102 to 6-103, McGraw-Hill, 1967.

5. U.S. Department of Energy Handbook 3010-94, "Airborne Release Fractions/Rates and Respirable Fractions for Nonreactor Nuclear Facilities," U.S. Department of Energy, Washington, DC, December 1994.

6. S.G. Homann, "HOTSPOT Health Physics Codes for the PC," Lawrence Livermore National Laboratory, UCRL-MA-106315, 1994.